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Development of a Ceramic Seal Handheld Reader and Fluorescent Seal Coatings

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Abstract
Savannah River National Laboratory (SRNL) and Sandia National Laboratories (SNL) are collaborating on development of a Ceramic Seal, which is a tamper indicating seal for potential arms control and international safeguards applications. As part of this research and development effort, SRNL is developing a handheld seal reader. The Ceramic Seal Reader (CSR) includes an electro-mechanical interface to the Ceramic Seal in order to read information from the seal. The Ceramic Seal contains a microcontroller chip that can detect tamper attempts. The reader is a battery powered hand held device designed to query the seal to determine its health and tamper status. A proof-of-concept reader is being developed and fabricated this year, with future developments to include data encryption and authentication. SRNL is also developing inorganic fluorescent coatings as a seal tamper indicating feature. The coating research and development is being performed in collaboration with small business partner Tetramer Technologies.

Introduction
Tamper indicating seals are used in nuclear verification regimes to verify that material is not diverted from a container or that unattended monitoring equipment is not tampered with. A seal must be reliable, cost-effective, easy to use, and must provide both tamper indication and unique identification (ID). In-situ verification of seals is also a desirable feature.[1]

The Ceramic Seal presents a new concept in low cost seal design. This seal brings together a number of technologies enabling a level of security that has previously been unavailable for applications where metal loop seals are currently used. Following are some of the significant improvements over previous passive seals:

- Frangible ceramic seal body
- Functional coatings to identify drilling and cutting attacks
- Intrinsic unique identifier using Laser Surface Authentication
- Electronics internal to the seal that provide in-situ cryptographic seal authentication and tamper indication
- User-friendly seal wire capture

An alumina ceramic base material for the seal body was selected to present a strong but brittle/frangible structure that will break rather than open after installation. Alumina ceramics are difficult to repair without leaving some telltale indication.

SRNL is performing this research and development in collaboration with Sandia National Laboratories (SNL). The design of the Ceramic Seal has gone through three major iterations, with Vulnerability Reviews performed by SNL throughout the process. Figure 1 shows the Generation I Ceramic Seal.

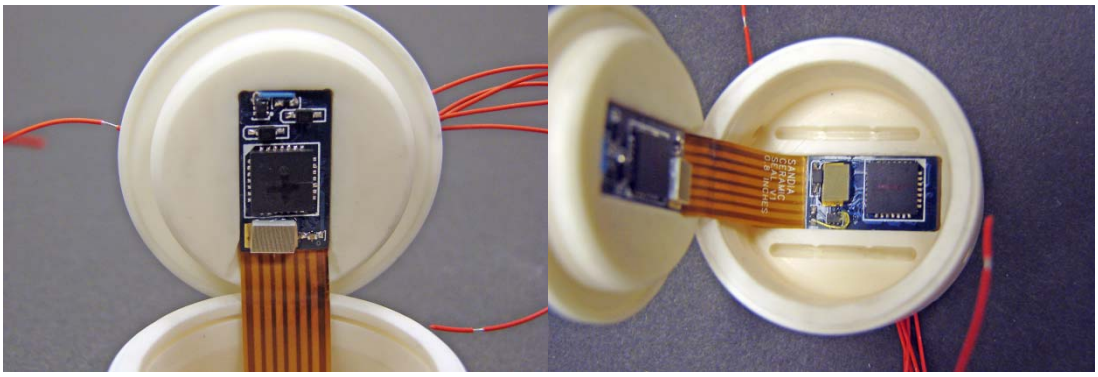

Figure 1: Generation I Ceramic Seal Assembly and Electronics (photos provided by SNL)

The design of the alumina body was developed by SRNL and the electronics were developed by SNL. The Generation II Ceramic Seal (see Figure 2) was fabricated by SNL using a layered manufacturing process called Low Temperature Co-Fired Ceramic (LTCC). The LTCC material is an alumina based ceramic that is built up in layers prior to firing. The LTCC body allows passive electronics and tamper indicating features to be incorporated into the seal body. Another change in the seal is improved seal wire routing, which retains the seal wire more securely and allows the wire to be installed without an installation tool.

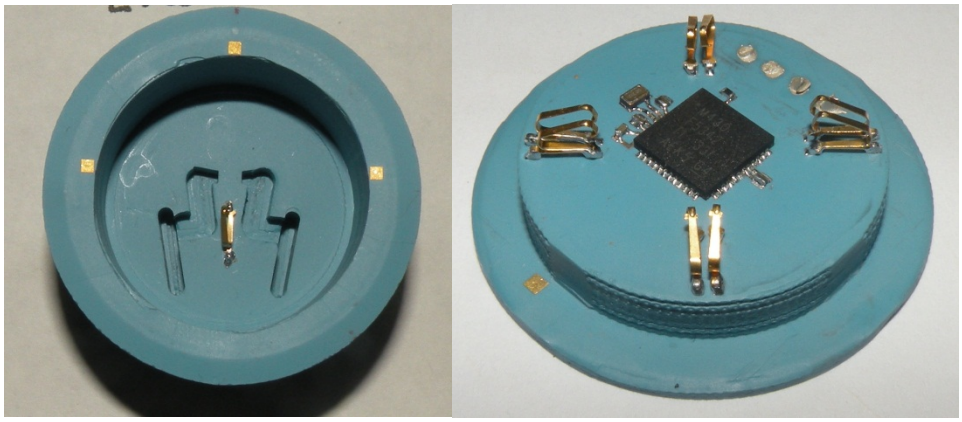

Figure 2: Generation II Ceramic Seal

Difficulties with implementing the complex features of the Ceramic Seal using the LTCC process and also the high cost of LTCC manufacturing and materials led the team back to alumina as the seal body material. The Generation III Ceramic Seal design is near completion and is to be fabricated out of the original alumina material (see Figure 3). Use of the alumina material allows the rest of the mechanical improvements intended for Generation II to be implemented. An indexing feature was added to the interface between the cap and base to ensure rotational alignment between the seal components is maintained while the seal is installed. An external flat was added to facilitate alignment for Laser Surface Authentication. Tamper planes are being added to the internal surface of the seal in lieu of the LTCC embedded tamper planes. The circuit board was revised and improvements were made to the electrical connectivity between the cap and the base.

Figure 3: Generation III Ceramic Seal (courtesy SNL)

Development of a Handheld Seal Reader

One of the advantages of the Ceramic Seal over existing low cost seals such as the metal seal is the ability to authenticate the seal and detect tamper attempts in situ. In order to achieve this capability, a handheld seal reader is being developed that will allow an inspector or arms control verifier to interrogate Ceramic Seals while the seals are installed on a component. The cap of the Ceramic Seal has external contacts that allow communication with the electronics internal to the seal. The initial reader concept was to design a custom reader case around an RLC embedded computer that uses the Microsoft Windows CE operating system. The reader case would have a cavity for inserting a Ceramic Seal for interrogation. RLC computers have been used successfully by SRNL for multiple previous applications, but SRNL was recently informed that RLC is going out of business and will no longer provide technical support for their computers.

Figure 4: RLC Computer

Figure 5: 3D Model of RLC Reader Concept

To avoid using an obsolete computer with no vendor technical support, a search was performed to determine the available options. One of the disadvantages of the RLC computer was that it required a voltage of at least 7.2 volt DC. This precluded using the small form factor, high power density batteries used in cell phones and other portable devices, which typically supply 4

volts or less, unless the batteries were configured in a series circuit. No desirable direct replacement was found for the RLC computer. However, a number of vendors were found that manufacture rugged handheld computers for industrial applications. Among the computers found, the Archer 2 from Juniper Systems (see Figure 6) was determined to have the best features for our application. It is designed for industrial use that is shock-resistant, waterproof, has a -22°F to 140°F operating range and has a high visibility screen for outdoor applications. The Archer 2 has the requisite computing power and multiple communications ports, including RS232, USB host and USB client. It also can operate for up to 20 hours on one charge. The Archer 2 uses the Microsoft Windows Embedded Handheld operating system.

Figure 6: Archer 2 Handheld Computer from Juniper Systems

Rather than modifying the rugged Archer 2 case, it was decided to design a separate seal module to connect to the Ceramic Seal electrical contacts. This module will communicate with the Archer 2 via USB. A 3D model of the Archer 2 and seal module is shown below in Figure 7. To use the seal reader, the seal would be slid into the seal module. The seal module is designed to automatically capture the seal and make electrical contact with the seal once the seal is fully inserted into the module. The module has internal electronics that alert the Archer 2 that a seal is connected. The Archer 2 then processes the data from the seal. If the state of health of the seal is “healthy” and no tamper attempts are registered, the seal module will indicate that the inspection is complete. If the seal is “unhealthy”, a tamper attempt is detected, or if there is an error in communicating with the module, the Archer 2 to will direct the seal module to indicate to the inspector that further information is needed. The inspector will then activate the Archer 2 to determine the path forward based on the information processed by the reader. Once the seal interrogation is complete, a lever on the seal module is pushed to eject the seal from the module.

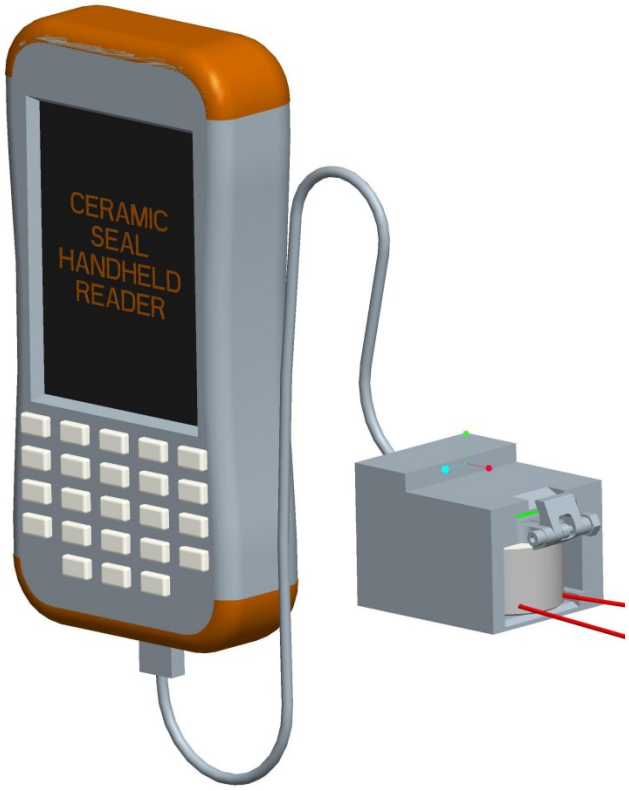

Figure 7: 3D Model of Archer 2 Handheld Reader with Ceramic Seal Inserted in Seal Module

Development of Fluorescent Seal Coatings

SRNL has researched functional coatings both for protection of the exterior ceramic surface for Laser Surface Authentication (LSA) and for tamper indication. LSA reads the reflections of a red laser off of the surface of the ceramic seal to develop a unique ID of the seal surface. Transparency of the coating in the red visible light wavelength range is required for the coating to be compatible with LSA. Both fluorescent and electrically conductive coatings have been investigated for tamper indication.

In previous years, SRNL developed various fluorescent coating formulations for application to the external surfaces of the Ceramic Seal. Among the coatings developed were alumina and silica sol gels and sputtered Indium Tin Oxide (ITO). ITO was initially considered due to its conductive properties as another potential tamper indication. It was decided that measurement of the surface conductivity as a tamper indicating feature would not be reliable because the conductivity of the seal coating would almost certainly be modified by wear and tear in an industrial environment. ITO was abandoned for this reason and also because sputtering is expensive relative to other processes.

Various fluorescent dopants were added to the sol gels to optimize luminescence. The combination of alumina sol gel with terbium dopant was found to offer the best stability and luminescence when applied to alumina ceramic substrates. SRNL used spin coating to apply the sol gels to flat alumina substrates.

During 2013, SRNL contracted Tetramer Technologies to develop a spray coating technique for application of tamper indicating coatings on the exterior of the Ceramic Seal. Specifically, Tetramer was contracted to apply alumina based solutions with two fluorescent dopant options and to demonstrate 3D application to the external surfaces of ceramic substrates. The coatings were specified to exhibit no obvious signs of cracking under normal lighting or when subjected to UV illumination and to have a thickness of at least one micron. The alumina sol gel formula that SRNL developed and used for spin coating application on flat alumina surfaces was provided to Tetramer for adaptation to the spray coating process. SRNL also provided alumina and LTCC substrates and seals for application of the coatings.

Initial work by Tetramer involved adapting the alumina sol gel formula developed by SRNL into a solution that could be applied using the Tetramer spray coating process. Referring to Figure 8, Variant 1 is a Tb/Al_2O_3 sol gel using Tb:acetate for the fluorescent dopant and Variant 2 is a Tb/Al_2O_3 sol gel using Tb nanoparticles for the fluorescent dopant.[2,3] The seal bodies in the top photos of Figure 8 are red due to the luminescence of the chromium content in the seal body itself. The coatings on the bottom photos of Figure 8 exhibited fluorescence after application of a proprietary oil treatment by Tetramer.

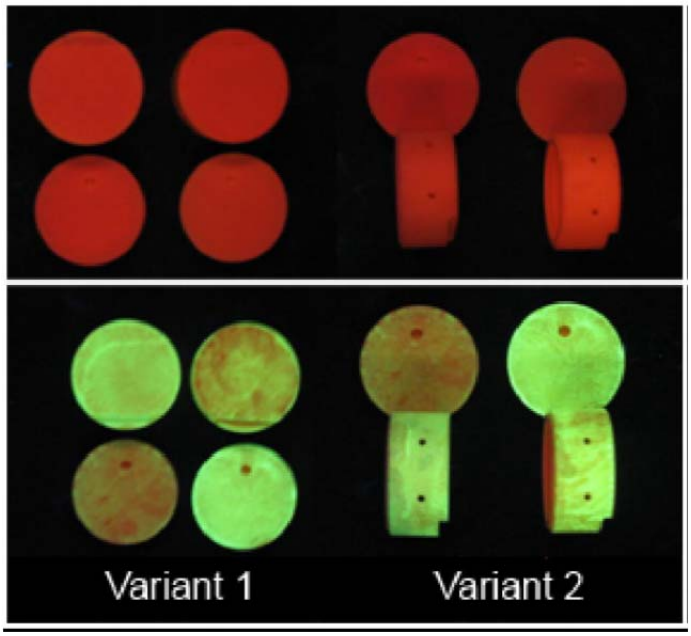

Figure 8: Alumina Ceramic Seal Bodies Under 254 nm UV Source Before (top) and After (bottom) Tetramer Treatment (courtesy Tetramer Technologies)

When it was decided to use LTCC as the seal body material, LTCC substrates began to be used by SRNL and Tetramer for coatings trials. Variant 1 was found to have better fluorescence and stability with LTCC substrates, so the seal bases and caps shown in Figure 9 are coated with Variant 1.

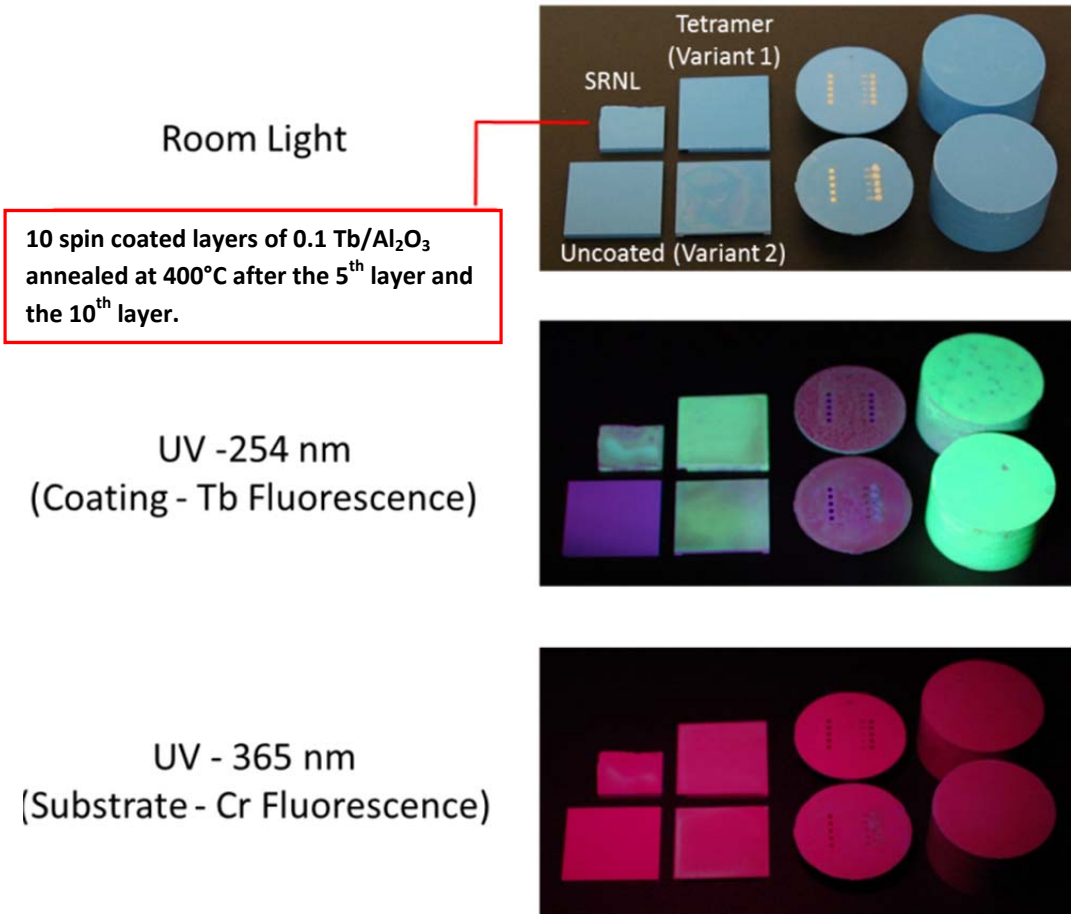

Figure 9: LTCC ceramic substrates and seal bodies under room and UV light.

Characterization of the initial coated substrates revealed that the more uniform coatings without cracks exhibited significantly less fluorescence than coatings with crack formations. In order to address this problem, Tetramer applied a propriety fluorescence enhancing additive after coating application and annealing (Figure 10).

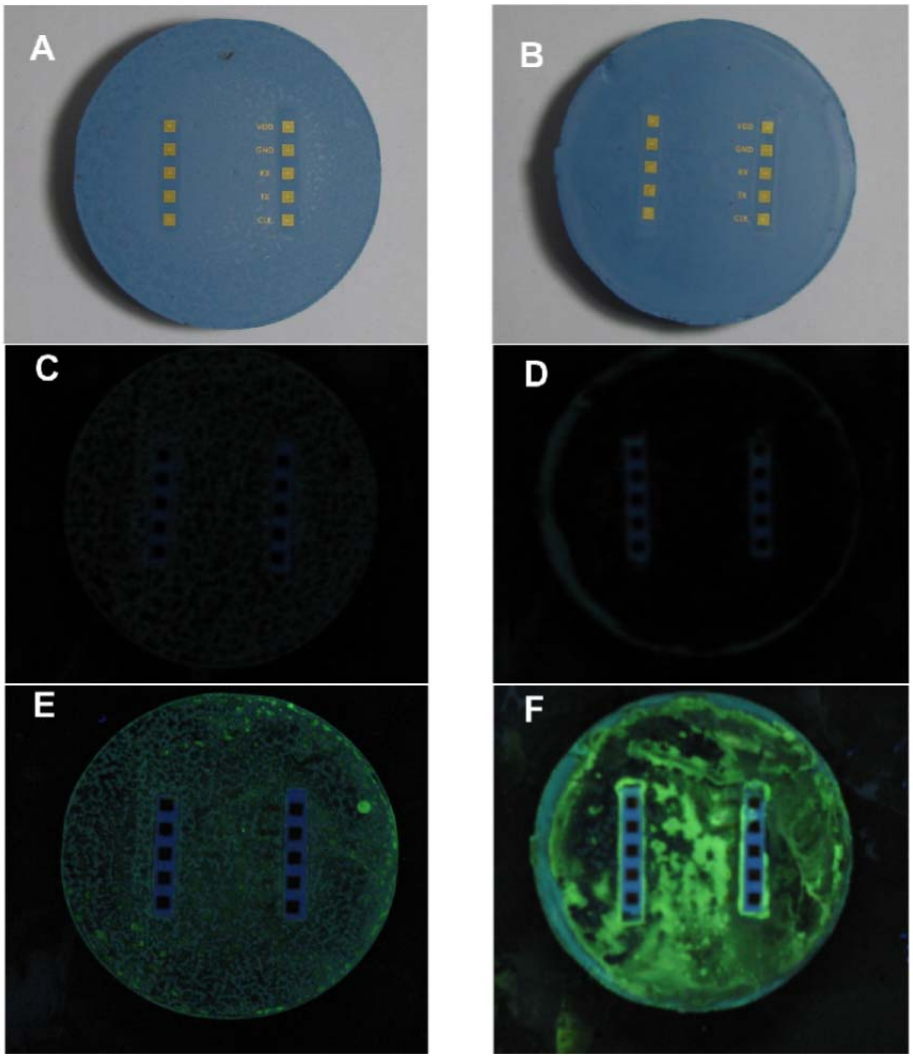

Figure 10: LTCC seal cap 1 (A, C, E) and LTCC seal cap 2 (B, D, F) after masking/coating/ annealing. Inspected in ambient indoor light (A, B), with a 254 nm UV light (C, D) and with the 254 nm light after the fluorescence enhancement additive was applied (E, F). (Courtesy Tetramer Technologies)

Figure 10 shows two LTCC seal caps after coating and annealing. Photos C-F demonstrate the increase in fluorescence caused by the fluorescence enhancement additive. When exposed to 254 nm UV light, the seal caps can barely be seen with the coating only (C, D), but the coating is easily visible after application of the fluorescence enhancement additive (E, F). The blue areas around the electrical contacts in photos E and F illustrate the ability to mask over areas of the surface that should not be coated. The mottled appearance of seal cap 1 is attributed to high viscosity sol gel, which has been addressed. The non-uniform appearance of seal cap 2 is attributed partly to the masks preventing uniform flow of the coating and partly due to non-uniform application of the fluorescence enhancement additive.

The LTCC seal bases were coated subsequent to the coating of the caps. Figure 11 shows the improvement of the coating on the bases relative to the caps, both in fluorescence and uniformity. The improvements are attributed to better control of the sol gel viscosity and improved fluorescent enhancement additive material and application method. The improved

uniformity is also due to the fact that no masking was required on the bases. More development is needed to ensure that uniform coatings can be applied in conjunction with masking. The spots on the coated bases are due to divots and imperfections in the surface of the LTCC material. The roughness of the cylindrical sides of the bases contributed to coating non-uniformity.

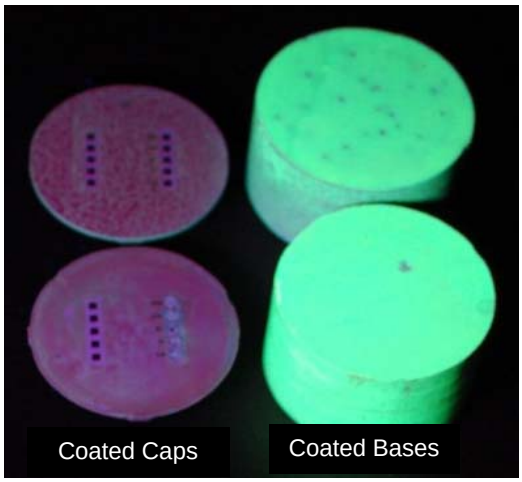

Figure 11: Comparison of Coating on Bases vs. Caps

Since the decision has been made to return the alumina ceramic seal material, no seals have been completed for application of a fluorescent coating. However, in addition to the coatings work Tetramer Technologies is performing for SRNL, they have received an NA-22 SBIR grant for development of polymer based tamper indicating coatings. The photos below illustrate promising polymer coatings that they have developed. The coated alumina substrates in Figure 12 are excited at 254 nm (top) and 365 nm (bottom). The left side is a graded coating that would be difficult to repair after a tamper attempt and the right side is a fluorescent Tetramer logo on an alumina substrate.

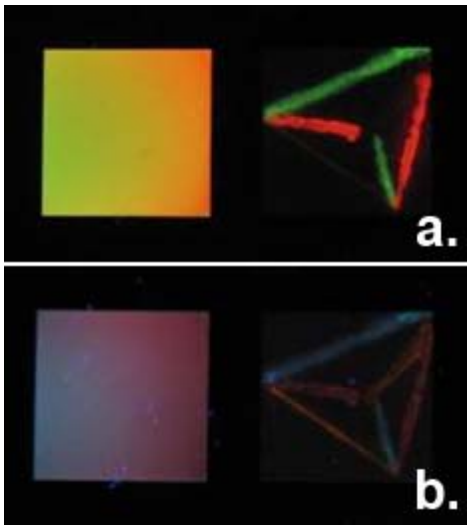

Figure 12: Potential Tamper Indicating Coatings

The coated alumina substrates in Figure 13 illustrate coatings that could potentially be used for seal authentication. The top photos illustrate a randomly generated pattern that is transparent in

visible light but could be used as an authenticating measure when excited by 254 nm UV using flicker comparison. The bottom photos show a 3D barcode that is only visible under 254 nm UV.

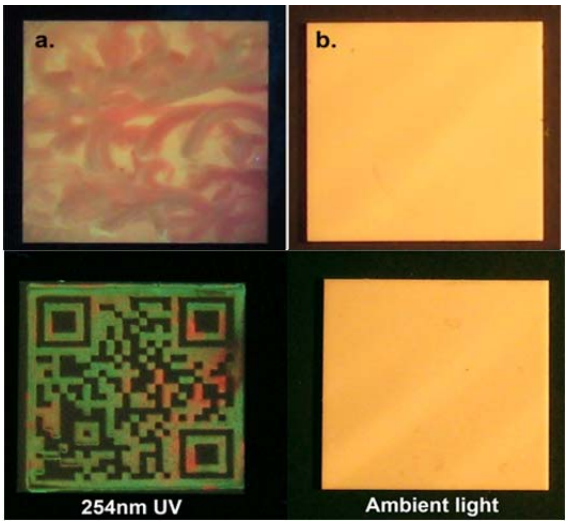

Figure 13: Potential Authenticating Coatings

Conclusion

The design of the Gen III Ceramic Seal is almost complete. Multiple seals will be fabricated and coated by Tetramer Technologies for honest game testing and field testing. This testing will be used to recommend improvements to the seal for commercialization. The phase one handheld seal reader design and fabrication will be completed in the next several months and testing of the reader will inform improvements to the phase two reader. In addition, a CRADA is being established to further the collaboration between SRNL and Tetramer Technologies, to include development of a portable coatings reader.

Acknowledgement

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